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#12

Attorney Docket No.: 003364P035

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Application of:

Il-Ki Woo, et al.

Serial No.: 09/494,211

Filed: January 25, 2000

For: LITHIUM SECONDARY BATTERY

Examiner: Dove, T.

Art Unit: 1745

DECLARATION PURSUANT TO 37 C.F.R. § 1.131

Honorable Commissioner  
for Patents  
Washington, D.C. 20231

Dear Sir:

**RECEIVED**

**JUN 21 2002**

**TC 1700**

I, Sang-Won Lee, hereby declare that:

1. I am a citizen of Korea.

2. I currently reside at San 24-1, Seongseong-dong, Cheonan-si,

Chungcheongnam-do, Korea.

3. I received a bachelor's degree in metallurgical engineering from Korea University in 1990 and a master's degree in materials science & engineering from Korea Advanced Institute of Science and Technology in 1992.

4. I am currently an employee of Samsung SDI Co., Ltd., Suwon-Si, Kyungki-do, Korea.

5. I have been employed by Samsung from 1992 to present.

6. My current title at Samsung is Senior Researcher.

7. I am a co-inventor of the above-identified patent application.
8. Samsung is the assignee of the above-identified patent application.
9. I have reviewed U.S. Patent No. 6,235,427 issued to Idota et al. ("Idota")


which was filed on May 11, 1999. The Examiner has cited Idota against the claims of the above-identified application.

10. The invention disclosed and claimed in the above-identified patent application was conceived in Korea at least as early as March, 1999, as evidenced by the Application No. 99-2257 filed in the Korean Industrial Property Office on January 25, 1999 (a certified copy of which was filed with the United States Patent & Trademark Office on September 13, 2001), and a translation of which is being filed herewith); my e-mail sent March 8, 1999 (a copy of which and a translation of which are being filed herewith); and Application No. 99-51148 filed in the Korean Industrial Property Office on November 17, 1999. The 99-2257 Application and my e-mail of March, 1999 were reduced to writing by Samsung at least as early as the date of my e-mail, i.e., March 8, 1999. These documents demonstrate a reduction to practice of the claimed invention of the instant application and were prepared under my direction and based on my own original work. Therefore, the reduction to practice of the invention disclosed and claimed in the above-identified patent application occurred prior to the filing date of Idota.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the above-identified application or any patent issued thereon.

Respectfully submitted,

Dated: June 4, 2002

  
Sang-Won Lee

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## Verification Statement For Translation

I, LEE, Hye-Sook, hereby declare that I am conversant in the Korean and the English languages and that I am the translator of the document attached and certify that to the best of my knowledge and belief the following is a true and correct English translation of the specification contained in Korean Patent Application No. 1999-2257.

Signature : *Hyesook Lee* LEE, Hye-Sook  
Date : April 8, 2002

KOREAN INTELLECTUAL PROPERTY OFFICE

This is to certify that the following application annexed hereto is a true copy from the records of the Korean Intellectual Property Office.

Application No.: Patent Application No. 1999-2257

Filing date: January 25, 1999

Applicant(s): SAMSUNG SDI CO., LTD.

This 20th day of January 2000

COMMISSIONER

(Translation)

APPLICATION FOR PATENT

RECEIPIENT: The Commissioner of the Korean Intellectual Property Office

FILING DATE: January 25, 1999

TITLE: LITHIUM SECONDARY BATTERY

APPLICANT(S):

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CODE NO.: 1-1998-001805-8

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CODE NO.: 9-1998-000023-8

GENERAL POWER OF ATTORNEY REGISTRATION NO.: 1999-000513-0

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CODE NO.: 9-1998-000453-2

GENERAL POWER OF ATTORNEY REGISTRATION NO.: 1999-000525-2

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REQUEST FOR EXAMINATION: YES

The above identified application and request for examination are duly submitted herewith pursuant to Articles 42 and 60, respectively, of the Patent Act.

OFFICIAL FEE(S):

FILING FEE: 29,000 Won

PRIORITY CLAIM: 0 Won

REQUEST FOR EXAMINATION: 173,000 Won

TOTAL: 202,000 Won

ATTACHMENT(S): 1 Copy of specification, claims, abstract (drawings)



## **ABSTRACT OF THE DISCLOSURE**

### **[ABSTRACT]**

A lithium secondary battery exhibiting good mechanical properties and using a thin negative current collector is provided. The lithium secondary battery includes a positive electrode formed by coating lithium metal oxides on a positive current collector and a negative electrode formed by coating carbonaceous materials or SnO<sub>2</sub> on a negative current collector. The negative current collector is made of a Cu-based alloy foil with a thickness of 20  $\mu\text{m}$  or less and the Cu-based alloy foil includes at least one material selected from the group consisting of nickel, titanium, magnesium, tin, zinc, boron, chromium, manganese, silicon, cobalt, iron, vanadium, aluminum, zirconium, niobium, phosphorous, bismuth, and misch metal. The lithium secondary battery further includes a separator interposed between the positive and negative electrodes and an electrolyte into which the positive and negative electrodes and the separator are immersed.

### **[KEY WORDS]**

lithium secondary battery, copper foil, nickel, magnesium, tin, zinc, current collector

## **[SPECIFICATION]**

### **[TITLE OF THE INVENTION]**

LITHIUM SECONDARY BATTERY

### **[DETAILED DESCRIPTION OF THE INVENTION]**

#### **[OBJECT OF THE INVENTION]**

#### **[TECHNICAL FIELD OF THE INVENTION AND RELATED ARTS]**

##### Field of the Invention

The present invention relates to a lithium secondary battery and, more particularly, to a lithium secondary battery which exhibits particular mechanical properties using a thin negative current collector.

##### Description of the Related Art

Conventionally, a copper foil with at least 99.8% purity has been used as a negative current collector for a lithium secondary battery. During charge and discharge, a negative electrode is expanded (an expansion of about 10%) and the expansion results in a tensile stress in a current collector made of the copper foil. However, as the durable tensile strength of copper foil is low, such a foil is easily torn.

The weak tensile strength of copper foil makes it difficult to reduce the thickness of the copper foil, which prohibits an increase in an amount of a negative active material in a battery. Accordingly, it is impossible to produce a battery having high capacity using copper foil.

#### **[TECHNICAL SUBJECT MATTER OF THE INVENTION]**

It is an object of the present invention to provide a lithium secondary battery exhibiting good tensile strength.

It is another object to provide a lithium secondary battery using a thin negative current collector.

5           These and other objects may be achieved by a lithium secondary battery including a positive electrode formed by coating lithium metal oxides on a positive current collector, and a negative electrode formed by coating carbonaceous materials or  $\text{SnO}_2$  on a negative current collector. The negative current collector is made of a Cu-based alloy foil with a thickness of 20  $\mu\text{m}$  or  
10 less and the Cu-based alloy foil includes at least one material selected from the group consisting of nickel, titanium, magnesium, tin, zinc, boron, chromium, manganese, silicon, cobalt, iron, vanadium, aluminum, zirconium, niobium, phosphorous, bismuth and misch metal. The lithium secondary battery further includes a separator interposed between the positive and negative electrodes  
15 and an electrolyte into which the positive and negative electrodes and the separator are immersed.

#### **[STRUCTURE AND OPERATION OF THE INVENTION]**

The present invention provides a lithium secondary battery exhibiting high capacity. The lithium secondary battery is manufactured by using a Cu-  
20 based alloy foil having both a good tensile strength and a thin thickness.

The Cu-based alloy foil includes at least one material selected from nickel, titanium, magnesium, tin, zinc, boron, chromium, manganese, silicon,

cobalt, iron, vanadium, aluminum, zirconium, niobium, phosphorous, bismuth, and misch metal. The amount of nickel is 0.8 to 4 wt% of the copper, that of titanium is 0.2 to 4 wt% of the copper, that of magnesium is 0.05 to 0.6 wt% of the copper, that of manganese is 0.1 to 1.0 wt% of the copper, that of zinc is 0.1 to 2.0 wt% of the copper, that of silicon is 0.1 to 0.5 wt% of the copper, that of phosphorous is 0.02 to 0.16 wt% of the copper, that of tin is 0.1 to 2.0 wt% of the copper, that of iron or cobalt is 0.01 to 2.0 wt% of the copper, and that of aluminum is 0.005 to 0.5 wt% of the copper. If the materials are out of this range, it is difficult to obtain a foil having the desirable tensile strength.

The Cu-based alloy foil is generally produced by an electro-plating process or a cold-rolling process.

In the battery of the present invention, the positive electrode may be produced by dissolving lithium metal oxides such as  $\text{LiCoO}_2$ ,  $\text{LiNiO}_2$ ,  $\text{LiMnO}_2$ ,  $\text{LiMn}_2\text{O}_4$  and a polyvinylidene fluoride binder in N-methyl pyrrolidone to make a slurry, coating the slurry on a positive collector made of aluminum foil, and drying the coated collector.

The negative electrode may be produced by dissolving an active material into or from which lithium ions are intercalated or deintercalated (i. e. carbonaceous materials such as crystalline carbon or amorphous carbon, or  $\text{SnO}_2$ ) and a polyvinylidene binder in N-methyl pyrrolidone to make a slurry. The slurry is coated on a negative collector of the present invention and then dried. The negative collector preferably has a thickness of 20  $\mu\text{m}$  or less and the Cu-based alloy foil with about 15  $\mu\text{m}$  of thickness can be used for the

collector without the deterioration of mechanical properties, such as its tensile strength.

The separator may be a porous film made of polyethylene or polypropylene.

5 The electrolyte may be a 1M solution prepared by dissolving  $\text{LiPF}_6$ ,  $\text{LiAsF}_6$ ,  $\text{LiCF}_3\text{SO}_3$ ,  $\text{LiN}(\text{CF}_3\text{SO}_2)_3$ ,  $\text{LiBF}_6$ , or  $\text{LiClO}_4$  in an organic solvent. The organic solvent may be of cyclic carbonates such as propylene carbonate or ethylene carbonate, linear carbonates such as dimethyl carbonate, or diethyl carbonate, or a mixture thereof.

10 The following examples further illustrate the present invention.

#### Examples 1 to 4 and Comparative Examples 1 to 2

Foil types for negative current collectors were manufactured according to the compositions shown in Table 1. The electrolytic copper foil was used in Comparative example 1 and the rolled copper foil was used in Comparative  
15 example 2. The tensile strengths thereof were measured and the results are present in Table 1. In Table 1, "Com." refers to "comparative example".

Table 1

	Composition	Tensile strength [N/mm <sup>2</sup> ]
Example 1	Ni: 1.8 wt%, Ti: 1.1 wt%, Cu: balance	560
Example 2	Ni: 2.0 wt%, Ti: 0.9 wt%, Mg: 0.13 wt%, Cu: balance	620
Example 3	Ni: 2.0 wt%, Ti: 1.1 wt%, Mg: 0.29wt%, Mn: 0.52 wt%, Cu: balance	620
Example 4	Ni: 1.5 wt%, Ti: 0.9 wt%, Mg: 0.26 wt%, Zn: 0.20 wt%, Cu: balance	630

Comp. 1	Cu: at least 99.9 wt%	420
Comp. 2	Cu: at least 99.9 wt%	340

As shown in Table 1, the collectors of Examples 1 to 4 have superior tensile strength to those of Comparative Examples 1 to 2. The collector of Example 4 having 1.5 wt% of nickel, 0.9 wt% of titanium, 0.26 wt% of magnesium, and 0.20 wt% of zinc has the highest tensile strength.

#### **[EFFECT OF THE INVENTION]**

As described, the present invention provides a negative current collector exhibiting improved mechanical strength and thermal conductivity by adding materials such as nickel or titanium to copper. The negative current collector of the present invention exhibits good tensile strength such that a wide collector can be produced to improve workability, and the thickness of current collector can be reduced to increase the capacity of the battery.

While the present invention has been described in detail with reference to the preferred embodiments, those skilled in the art will appreciate that various modifications and substitutions can be made thereto without departing from the spirit and scope of the present invention as set forth in the appended claims.

**[WHAT IS CLAIMED IS:]**

1. A lithium secondary battery comprising:

a positive electrode formed by coating lithium metal oxides on a positive current collector;

5 a negative electrode formed by coating carbonaceous materials or  $\text{SnO}_2$  on a negative current collector; the negative current collector being made of a Cu-based alloy foil with a thickness of  $20\ \mu\text{m}$  or less and the Cu-based alloy foil including at least one material selected from the group consisting of nickel, titanium, magnesium, tin, zinc, boron, chromium, manganese, silicon, cobalt,  
10 iron, vanadium, aluminum, zirconium, niobium, phosphorous, bismuth, and misch metal;

a separator interposed between the positive and negative electrodes;  
and

an electrolyte into which the positive and negative electrodes and the  
15 separator are immersed.

2. The lithium secondary battery of claim 1 wherein the amount of nickel is 0.8 to 4 wt% of copper, the amount of titanium is 0.2 to 4 wt% of copper, the amount of magnesium is 0.05 to 0.6 wt% of copper, the amount of manganese is 0.1 to 1.0 wt% of copper, the amount of zinc is 0.1 to 2.0 wt% of  
20 copper, the amount of silicon is 0.1 to 0.5 wt% of copper, the amount of phosphorous is 0.02 to 0.16 wt% of copper, the amount of tin is 0.1 to 2.0 wt% of copper, the amount of iron or cobalt is 0.01 to 2.0 wt% of copper, and the amount of aluminum is 0.005 to 0.5 wt% of copper.